COMPARISON OF MICROWAVE-ASSISTED HYDRODISTILLATION WITH THE CONVENTIONAL HYDRODISTILLATION METHOD IN EXTRACTION OF ESSENTIAL OIL

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ABSTRACT

The Basil (*Ocimum basilicum*) essential oil was a significant interest as a new high-value essential oil especially in pharmaceutical, aromatherapy aid and cosmetics industries which give large opportunities for global marketing. Traditional extraction method Hydodistillation (HD) used to obtain essential oil have several drawbacks which are longer time consuming, have low extraction yields, and higher operational costing. At present the most appropriate technique to improve the quality of essential oil was by using Microwave assisted Hydrodistillation (MAHD) which able to overcome the drawbacks like mentioned above is introduced. This study obtained to identify the effect of the extraction time, yield and composition of Basil essential oil for HD and MAHD. Ratio of Basil and water used was 1:8 were placed in the HD and MAHD setup. The essential oil components were identified by using GCMS. The result shows MAHD can obtained higher yield at lowest extraction time due to more efficient heat transfer involved with microwave. The dominated compound was Methyl cinnamate (34.38% and 66.26%) an oxygenated compound for both HD and MAHD. As a conclusion MAHD obtained greater yield with shorter time and high percentage of oxygenated compounds compared with HD. Furthermore MAHD shows a good alternative method to produce essential oil of Basil.

ABSTRAK

Minyak asli Basil (*Ocimum basilicum*) adalah amat bermanfaat sebagai minyak asli baharu yang mendapat permintaan tinggi terutamanya dalam industri perubatan, aromaterapi dan kosmetik yang mempunyai nilai pasaran dunia. Kaedah penyulingan tradisional berasaskan air suling (HD) digunakan untuk menghasilkan minyak asli ini mempunyai beberapa kelemahan seperti jangka masa proses yang lama, kadar penghasilan produk yang rendah dan kos operasi yang tinggi. Pada masa kini teknik yang paling optimim untuk meningkatkan quality minyak asli adalah menggunakan Microwave sebagai sumber tenaga untuk penyulingan berasaskan air (MAHD) yang mampu mengatasi kelemahan-kelemahan yang dinyatakan di atas diperkenalkan. Kajian ini bertujuan untuk mengenalpasti kesan masa extrak, kadar penghasilan produk dan komposisi miyak asli Basil untuk kaedah HD dan MAHD. Nisbah kuantiti basil dan air yang digunakan adalah 1:8 diisi kedalam aturan HD dan MAHD. Komposisi minyak asli dikenalpasti dengan menggunakan alat aplikasi GCMS. Data yang diperolehi melalui MAHD menunjukkan kadar penghasilan minyak asli yang tinggi dalam masa yang singkat merujuk kepada pemindahan haba yang amat efisien menggunakan tenaga microwave. Komposisi dominan adalah Methyl cinnamate (34.38% and 66.26%) iaitu terdiri daripada komposisi ‘oxygenated’ untuk kedua-dua kaedah HD dan MAHD. Kesimpulannya MAHD menghasilkan kadar penghasilan produk yang tinggi dalam masa yang singkat dan peratusan komposisi ‘oxygenated’ yang tinggi berbanding kaedah HD. Oleh itu MAHD menunjukkan kaedah alternatif yang berpotensi untuk menghasilkan minyak asli Basil.

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US$	= Dollar (United State)
^{o}C	= Degree Celsius
%	= Percentage
kPa	= Kilo-Pascal
CO_2	= Carbon Dioxide
MHz	= Mega-Hertz
W	= Watts
e/m	= Mass Ratio
mL	= Mili-Liter
g	= Gram
L	= Liter
min	= Minutes
hr	= Hours
Hz	= Hertz
m	= Meter
mm	= Mili-Meter
cm/s	= Centi-Meter Per Second
$^{o}C/min$	= Degree Celcius Per Minutes
µL	= Micro-Liter
V	= Volume
V/W	= Volume/Weight

LIST OF ABBREVIATIONS

FRIM	= Forest Research Institute Of Malaysia
MARDI	= Malaysian Agricultural Research And Development Institute
HD	= Hydrodistillation
MAE	= Microwave-assisted Extraction
SFE	= Supercritical Fluid Extraction
MAHD	= Microwave-assisted Hydrodistillation
GCMS	= Gas Chromatography Mass Spectrometry
CNS	= Central Nervous System
WW I	= World War I
SC CO_2	= Supercritical Co_2
SE	= Solvent Extraction
SD	= Steam Distillation
GC (TD/GC)	= Gas Chromatography (Thermal Desorption / Gas Chromatography)
TD-GCMS	= Thermal Desorption- Gas Chromatography Mass Spectrometry

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Nowadays people looking towards new product base natural sources since there are no side effect to society and environment when taken accordingly. People interest in the production which have high value, good quality, environmentally friendly approached and low cost operation. Higher demanding with variety products vary from essential oil. Essential oil holds high export potential because of it properties which are natural volatile compounds from plant materials. The ordinary feature of essential oil possessed the essence of a plant, the particular aroma, flavor and other characteristic that was practical used such as perfumes, food flavors and pharmaceutical. Essential oils are among the secondary metabolites produced within the various organs of medicinal herbs/plants. They are complex mixtures of volatile compounds such as terpenes (mostly monoterpenes and sesquiterpenes), phenolics and alcohols (Lucchesi, *et al.*, 2004).

Due to the strong and sweet aroma present in *Cymbopogon citratus*, the economic importance of *Cymbopogon citratus* includes its use in aromatherapy. *Cymbopogon citratus* (lemon grass) both deodorizes and is an effective antiseptic. It is excellent for tired and aching feet. Lemon grass oil may be used in the treatment of acne, athlete's foot, excessive perspiration, flatulence, insect- repellant, muscle aches, oily skin, scabies and stress. Fresh basil (*Ocimum basilicum* L.) is used in food,

phytotherapic industry, and in traditional therapeutic, due to its essential oil content and composition. There is a significant interest in basil as a new high-value essential oil.

Besides of Basil and *Cymbopogon citratus* ginger also can be extract to essential oil. Most of the ginger oil base products are being imported from other countries due to lack of interest and technology in the third world countries and in the developing countries. Malaysia should be more actively involved in the production of essential oil. Due to lack of technology and knowledge of ginger oil and oleoresin extraction processes in Malaysia, much research is now being carried out to fulfill the demand of herbal base products in this region. In Malaysia, ginger oil extractions are mostly done by hydro-distillation. Local institutions like the Forest Research Institute of Malaysia (FRIM) and Malaysian Agricultural Research and Development Institute (MARDI) play a major role in the essential oil technology transfer. Most of the research published done on ginger oil locally and worldwide are on the chemistry part and lack in the processing part.

Due to the increasing of the essential oil demanding, many researchers develop various novel methods for extraction process such as hydrodistillation (HD), microwave-assisted extraction (MAE), steam distillation, solvent extraction, and supercritical fluid extraction (SFE). The most common approach to extract essential oils from the plant materials is HD which is conventional method where the essential oils are evaporated by heating the mixture of water and plant materials. Then the vapor in a condenser was liquefaction (Rezvanpanah, *et al*., 2008). This method have several disadvantages such as take a long extraction time, higher operational cost due to the long time process losses in the volatile compounds and degradation of some components through thermal effect.

The development of essential oil sector has a direct relation with the improvement of process technology in industrial plants. Hence in order to reduce the extraction time, operational cost, enhances the yield and quality of the essential oil an alternative approaches which is Microwave-assisted Hydrodistillation (MAHD) has been

developed. MAHD is an advanced hydrodistillation method base on the use of microwave oven to extract volatile materials. MAHD utilize heat transfer within the sample by three ways which is irradiation, conduction and convection. As a result, the heat produced is more quickly compare to HD methods which is only can transfer heat through conduction and convection only. The essential oil components were identified by comparing retention times of the chromatogram peaks with those of references compounds run under identical conditions by using GCMS.

1.1 Problem Statement

Higher demanding of essential oil as pharmaceutical, aromatherapy aid and cosmetics ingredients give large opportunities for global marketing. The worldwide market for essential oil growth rapidly. On top of that, nowadays a lot of scientific research presently focused on the industrial development together with environmental preservation. In this contact it is necessary to find the most appropriate technique or method to improve the quality of essential oil. Traditional extraction method used to obtain essential oil have several drawbacks which are longer time consuming, have low extraction yields, laborious, and higher operational costing. Furthermore this method utilizes large amounts of toxic solvent which will effects the environment.

At present, alternative extraction method which is able to overcome drawbacks like mentioned above by using MAHD method. This method provides higher selectivity; shorten extraction time, effectiveness cost, not use toxic organic solvent and environmentally safe technology. Therefore, MAHD technique is finding wider application in natural product separation. Microwave heating sources has an incontestable place in analytical and organic laboratory practices as an effective and non-polluting method. This method introduced as a safe method because it's does not involve in any deterioration of the extracted components.

1.2 Objectives

The objective of this study is to:-

I. Comparison of MAHD and HD method in extraction of essential oil (Basil and *Cymbopogon Citratus*) of their chemical composition.

II. Identify the effect of variation of extraction parameters such as extraction time and operational cost.

III. Investigate the effect of microwave energy on the yield or efficiency and quality of the essential oil against traditional method hydro distillation.

1.3 Scope Of Study

In order to achieve the objective, the following scopes have been identified:-

I. Study the effect of extraction time for an essential oil recovery. Different time will obtained for various method and MAHD need shorter times than HD.

II. Comparing the operational costing for MAHD and HD method. In order to save in the extraction cost, MAHD is the most preferred because it will save the electricity due to the short time process.

III. Comparing the extraction yield that will be achieved when using MAHD and HD.

IV. Analyze the composition of essential oil of sample by using Gas Chromatography-Mass Spectrometry.

CHAPTER 2

LITERATURE REVIEW

2.1 Essential Oil

An essential oil is a concentrated hydrophobic liquid containing volatile aroma compounds from plant. They are also known as aromatic oils, fragrant oils, steam-volatile oils, ethereal oils, or simply as the "oil of" the plant material from which they were extracted, such as *oil of clove*. The advantages of essential oils are their flavour concentrations and their similarity to their corresponding sources. The majorities of them is fairly stable (notable exception is the citrus oil) and contain a few natural antioxidants. Although most of the essential oil is soluble in high strength alcohol, it's had poor water solubility and most contain monoterpenes that contribute to their poor water solubility. Essential oils are among the secondary metabolites produced within the various organs of medicinal herbs/plants. They are complex mixtures of volatile compounds such as terpenes (mostly monoterpenes and sesquiterpenes), phenolics and alcohols (Lucchesi *et al.*, 2004). However, the essential oils are highly complex and may include oxygenated compounds.

Essential oil is used in perfumery, aromatherapy, cosmetics, incense, medicine, household cleaning products and for flavouring food and drink. They are valuable commodities in the fragrance and food industries. More than 250 types of essential oils (120000t world annual production) worth US$ 1.2 billion per annum are traded in the world market. A number of countries produce different kinds of essential oils. India ranks second in the world trade of essential oils (Rao, *et al*, 2005). Essential oil can be isolated using a number of isolation methods such as hydrodistillation (HD), steam distillation (SD), supercritical fluid extraction (SFE) and solvent extraction. Essential oil components were identified by comparing retention times of the chromatogram peaks with those of references compounds run under identical conditions, by comparison of retention indices where it were computed by logarithmic interpolation between *n*-alkanes (Kovats, 1965).

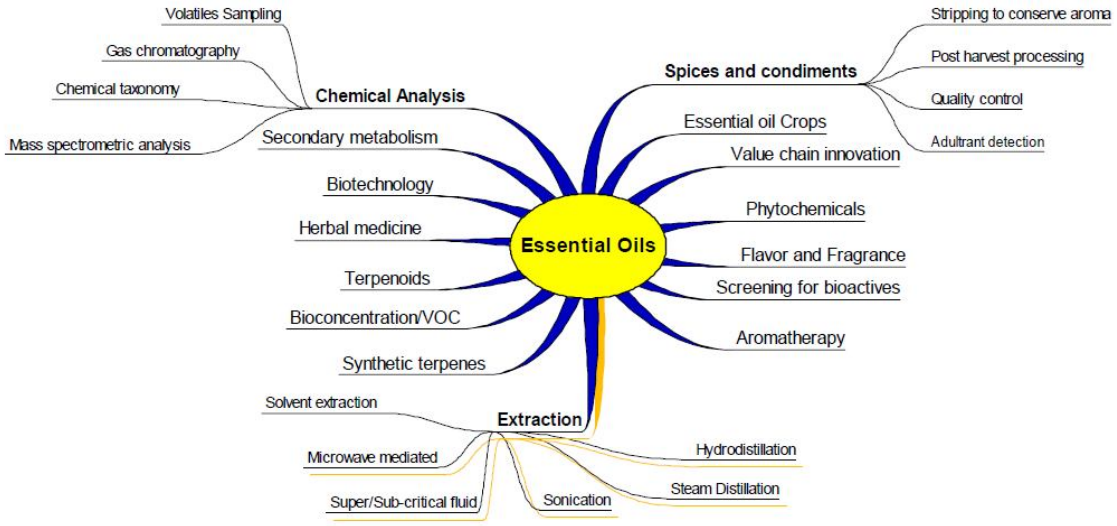

Figure 2.1: Tree diagram showing the wide branching of specializations in the field of essential oils

2.2 Others Raw Material of Essential Oil Extraction

2.2.1 Gaharu / Agarwood

Gaharu/Agarwood is the occasional product of two to four genera in the family *Thymelaeaceae*, with *Aquilaria agallocha, Aquilaria crassna* and *Aquilaria malaccensis* the best known species. The wood is formed as a result of the tree's immune response to fungal infection. The odor of agarwood is complex and pleasing, with few or no similar natural analogues. Gaharu contains a sesquiterpene alcohol which produces its characteristic aroma. The production of gaharu resins have a several potential such as used primarily in traditional Chinese and Korean medicines. The oil also used in perfumes and cosmetic product. Besides that the chips of agarwood are ground into powder can be used for special cigarettes. Its ethereal fragrance is demanding for incense which used in religious and spiritual ceremonies of Islam or Buddhism especially in Asia such as Thailand, Malaysia and Indonesia. Agarwood might have some effects towards central nervous system (CNS) such as higher brain function, from traditional use as a sedative (Ueda, *et al.*, 2006).

Differences in other chemical components were also noted between the best and lesser quality agarwood . As a result, agarwood and its essential oil gained great cultural and religious significance in ancient civilization around the world. Extraction of oil from agarwood can be determined through distillation process. This delicate process determines both the amount and quality of oil produced. In this process most of the wood is ground into very small pieces of powder as in Figure 2.2, which are immersed in water and left to ferment over time. This is due to the exception of large solid pieces of agarwood which are traded as individual pieces. Then the material is transferred to distillation kettles and steamed. After heating the condensed water and oil are captured in a container where the oil floats on top of the water. The water is removed and the oil is tapped.

Figure 2.2: Grinding Agarwood

2.2.2 Lavender

The Lavenders *Lavandula* are a genus of 39 species of flowering plants in the mint family, Lamiaceae, native to the Mediterranean region south to tropical Africa and to the southeast regions of India. The genus includes annuals, herbaceous plants, subshrubs, and small shrubs. The native range extends across the Canary Islands, North and East Africa, south Europe and the Mediterranean, Arabia, and India. Because the cultivated forms are planted in gardens world-wide, they are occasionally found growing wild, as garden escapees.

Lavenders are widely grown in gardens. Flower spikes are used for dried flower arrangements as in figure 2.3. The fragrant, pale purple flowers and flower buds are used in potpourris. Dried and sealed in pouches, they are placed among stored items of clothing to give a fresh fragrance and as a deterrent to moths. The plant is also grown commercially for extraction of lavender oil from the flowers. This oil is used as an antiseptic and for aromatherapy.

In medicinal plants, Essential oil of lavender has antiseptic and anti-inflammatory properties. It was used in hospitals during World War I (WWI) to

disinfect floors and walls. Lavender essential oil has a calming scent which makes it an excellent tonic for the nerves and helps in treating migraines, headaches, anxiety, depression, nervous tension and emotional stress. It's refreshing aroma removes nervous exhaustion and restlessness and increases mental activity. These extracts are also popularly used as fragrances for bath products. Lavender is also used extensively as herbal filler inside Sachets used freshen linens and discourage moths from closets and drawers. Dried lavender flowers have also become recently popular used as confetti for tossing after a wedding. In the food manufacturing, lavender essential oil is employed in flavoring beverages, ice cream, candy, baked goods and chewing gum.

Figure 2.3: Lavender

2.2.3 Ginger

Ginger or its scientific name is *Zingiber officinale* Roscoe is another aromatic plant which has been higher demanding due to its multiple functional activities. Ginger is monocotyledonous plant belonging to the family of *Zingiberacea*. The rhizome contains both the flavor and pungency of the spice or oleoresin together with essential oils. The peculiar hot taste and pungent taste of ginger can be attributed to the presence of an acrid compound called Gingerol. Most of the health benefits of ginger are due to Gingerol.

Ginger is a commonly used spice, which has originated in India as shown in figure 2.4. It forms an integral part of many Asian cuisines due to its digestive properties. It is especially helpful in digesting food items such as meat, and poultry and is added while cooking meat as it softens the meat. Ginger root and ginger oil are also used as preservative and flavoring agent. The health benefits of ginger root oil can be attributed to its digestive, carminative, expectorant, antiseptic, analgesic, anti inflammatory, stimulating and aphrodisiac properties. These benefits of ginger oil include its ability to treat stomach problems, nausea, heart strokes, indigestion, inflammations, respiratory problems, menstrual disorders.

However the detail information on the processing of ginger oil locally and worldwide is not enough. Most of the documented information was on the chemistry part, methods and the equipment used for essential oil extraction. Hence difficulty occurred for obtaining data specifically for ginger oil production.

Figure 2.4: Ginger

2.2.4 Basil (*Ocimum basilicum*)

The genus *Ocimum* L. has varieties of species, with a great variation in phenotype, oil content, composition, and possibly bioactivity which plays a major role in the Italian and Southeast Asian cuisines of Thailand, Vietnam, Cambodia and Loas. Holy basil [*Ocimum sanctum* L. (syn. *Ocimum tenuiflorum* L.)] and sweet basil (*Ocimum basilicum* L.) are the two basil species that are considered to be promising essential oil crops. The basil essential oil contains pleasant aroma and is known to possess antimicrobial, insecticidal, antiviral, anti-cancer and antioxidant activities. Due to its pleasant aroma and antimicrobial activity, basil essential oil is a major aromatic agent with applications in various industries such as the food, pharmaceutical, cosmetic, and aromatherapy industries.

The use of fresh basil (*Ocimum basilicum*) in food, phytotherapic industry, and in traditional therapeutic is frequently reported. This interest is due to the presence of essential oil, whose main components can be eugenol, methyleugenol, linalool, methylchavicol, or 1,8-cineole, depending on factors as chemotype, growth conditions and plant developing stage (Vieira & Simon, 2000). Certain basil essential oils were claimed to have a larvicidal activity towards mosquito's larvae. Azhari et al (2009) indicated that basil essential oils have larvicidal activity towards *Anopheles* larvae. Therefore, could be affordable way to control this mosquito.

The traditional and most widely used method for basil extraction is via steam distillation of the whole aboveground basil herbage (stems, leaves, and flowers). When grown for essential oil production, basil is harvested in full bloom, because the content and the composition of the oil are optimal at that stage. Depending on the climate, basil could be harvested one to three times during the cropping season (Topalov *et al*,. 1962). A different chemotype, the Reunion basil, is characterized by high levels of methyl-chavicol, whereas the tropical chemotype of basil is known to have methyl cinnamate as the main component of the essential oil. Another basil chemotype that is high in eugenol is grown in North Africa, Russia, Eastern Europe, and parts of Asia (Marotti *et*